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On depolarisation in 0D systems: Lamb-like level shift

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Abstract. An extremely quantized 0D system is studied for better understanding of an importance of many-body corrections to an one-electron spectrum. A new approach is proposed to describe the correction appearing as a depolarisation level shift of a (confined) electron moving within an electric field of a (confined) collective mode. For a quantum dot the correction diminishes at the size about 10^4-10^5 atoms, depending also on other system parameters. For a closed-shell carbon cluster the effect does not depend on the cluster size owing to stronger quantization and the one-electron estimation does not fit anywhere. For the semiconductor quantum dot system an experimental method to check the depolarisation model is proposed.

In the paper we study an anomalous large level shift, resulted from the interaction of an electron in a 0D-system with zero-point oscillations of confined modes of the electric field. Of course, any complete many-body theory, taking into account all the Coulomb interaction, gives the correct value for the electron levels, though it is not known for present. We go to reveal an important correction treating the effect of valence electrons of a 0D object, which can be a spherical quantum dot and a closed-shell fullerene cluster, selfconsistently. Then the theory remains to be semi-classical while a nature of the effect is quantum-electrodynamical. This continues our consideration of C_{60} in frame of a simple quantum mechanical model of the spherical-shell quantum well (SSQW) [1].

The energy correction depends on the system radius. This size scaling of the depolarisation is computed within an approach proposed by Migdal [2] for a calculation of Lamb shift in a hydrogen-like atom. The shift of the one-electron level is quite predictable and we will show that its amount becomes very large for the quantized system. The closed-shell fullerene depolarisation is of the order of the bare energy and independent of the cluster radius while the shift in the quantum dot decreases with the increasing number of atoms. Between two examples — a fullerene carbon nanocluster and a semiconductor quantum dot structure — the latter has not only theoretical importance. The possible experimental manifestation of the depolarisation effect is proposed basing on the spectroscopy of the quantum dot levels for different matrix materials.

1. Theory for depolarisation level shift: C_{60}

1. The use of the group of full rotations, SO(3), allows one to label the one-electron states and to get analytically the solution for the selfconsistent RPA response function of C_{60} [1]. A peak of a collective excitation shows up in this spectrum, resulting from fast coherent oscillations of a total electron density of valence states. This surface density oscillation can be thought as a confined electrical field mode or the surface plasmon.

We have considered semiclassically the LS for an arbitrary shell object in [3], followed to Migdal [2]. The frequency of the zero-point fluctuations of the external field is much

higher than the inverse period of the electron orbit $\omega_p \gg \pi/\tau$. Therefore, the adiabatic approximation has to be used and one divides the fast (field) and slow (electron) variables. An electron is subjected to short fast deflections from its original orbit in the high-frequency field of the electromagnetic wave of the zero-point fluctuation. Then the energy shift is given by the second order perturbation theory as

$$\delta E = \langle H(r+\delta) - H(r)\rangle = \left\langle \nabla H \cdot \vec{\delta} + \frac{1}{2}\nabla^2 H\, \vec{\delta}\cdot\vec{\delta} + \ldots \right\rangle = \frac{1}{4}\nabla^2 H\, \overline{\delta^2} + o(\overline{\delta^2}) \quad (1)$$

where $H(r)$ is the unperturbed Hamiltonian and $H(r+\delta)$ is the Hamiltonian with account for the random electron deflection δ. The angle brackets represent the quantum mechanical average over the fast variables of the field (or, the same, over the random electron deflections). The perturbed Hamiltonian is expanded in series on the δ and a first nonzero contribution is taken.

The expression for the mean square of the deflection, $\overline{\delta^2}$ was deduced in Ref. [3]. Though the estimation is semiquantitative, the deflection is of the order of atomic unit, $a_B \simeq 0.53$ Å. The $\overline{\delta^2}$ in the SSQW does not depend on the radius, neither on the number of atoms because of the density of the valence electrons is the same.

Let suppose that one-electron model works for some cluster C_N. The one-electron Hamiltonian reads as [4]:

$$H_o = E_n + \hbar^2/2mR^2\ \hat{L}^2, \quad (2)$$

where E_n is the energy of a lowest level of n-th radial series; an orbital quantization energy $\hbar^2/mR^2$ defines the SO(3) level spacing between eigenstates of the angular momentum operator. The SSQW level shift reads as follows:

$$E_L = E_L^{(0)}\left(1 + \kappa\hat{L}^2/N\right), \quad (3)$$

where $\kappa \sim 0.36$ is the numerical coefficient depending only on b, the carbon bond length: $\kappa = \sqrt{a_B/b}\pi^2/2^{2.5}/3$.

2. Within the closed-shell model the optical gap occurs between the levels $|L_F\rangle$ and $|L_F+1\rangle$. Within the closed-shell approximation the Fermi momentum fulfills the condition $N = 2\sum_{L=0}^{L_F}(2L+1) = 2(L_F+1)^2$. The gap value does depend on the cluster size, decreasing to the zero as N going to infinity in order to approach the gapless graphite.

The depolarisation makes the gap wider. The renormalisation is universal for any closed-shell spherical cluster and amounts about 40% to the bare value:

$$E_g = E_g^{(0)}(1+\kappa) \simeq 1.36E_g^{(0)}. \quad (4)$$

where the parameter $\kappa \simeq 0.36$ is the same as before.

2. Depolarisation energy level shift in QD structure

1. In order to evaluate the LS for the quantum dot (QD) the simplified *spherical* model in frame of an *effective mass* approximation was applied. The size scaling of the depolarisation shift is not sensitive to the model used, being dependent mainly on the corresponding density of states of the collective modes.

The simplest QD Hamiltonian is considered to have only the rotational correction which reads as:

$$\delta H = \frac{\hat{L}^2}{2mR^2}\left(-2\frac{\delta}{R} + 3\frac{\delta^2}{R^2} + \ldots\right) \quad (5)$$

where R is about the spherical QD radius; m is the electron mass which is supposed to be constant within the dot; $\hat{L}$ is the angular momentum operator.

Nearly self-evidently the bulk plasmon shift is negligible. The small factor, contained in the 3D LS, comes essentially from the expression for $\overline{\delta^2}$ which scales as $1/N$ [3], where N is the number of atoms in the QD. The mean square deflection, caused by the 3D mode (which is not confined at all), decreases with N too rapidly.

The square of the deflection [3] $\overline{\delta^2} = e^2/4m^2 \int d^3k \ \mathcal{E}_k^2/\omega_k^4$ is proportional to the square of the electric field strength. The field strength can be rewritten as the zero-point oscillation frequency $\mathcal{E}_k^2 = 2\pi\hbar\omega_k$ through the quantized field normalisation. The 3D plasmon frequency does not depend on the quantum number $\mathbf{k}$. Hence, the mean square deflection contains the total number of states effecting on the electron level in the QD. The integral is limited above by $k_{\max} \sim 1/R$. In 3D-case it brings the factor $R^{-3} \sim N^{-1}$ claimed in the beginning of the section. Then the depolarisation level shift due to 3D modes scales with N as follows:

$$\Delta_{3D} = \frac{\delta E}{E^{(0)}} \propto N^{-5/3}. \tag{6}$$

The rude estimation of the prefactor shows that even for the small QD with $N = 100$ the shift is 10^{-6} of the bare energy and will not be resolved because of a number of other different factors effecting the level position.

To give a complete picture, the standard LS due to the zero-point oscillations of the free electromagnetic modes of the vacuum reads as follows: $\Delta_{\text{vac}} \propto \alpha^3 N^{-2/3}$, where $\alpha \simeq 1/137$ is the fine structure constant. Though the slope of the LS in N is much slower than in Eq. (6) the prefactor is tiny because of α^3.

2. Two possible candidates for the confined plasmon modes in the QD system are the 2D plasmon and the 0D spherical mode. The former mode can arise because of some interface possibly grown within the structure (see inset in Fig. 1). It might be a wetting layer, if it is thick enough to confine the electromagnetic field. The 2D plasmon naturally originates at the boundary between the semiconductor structure and a distinct substrate. The scaling in N will have a lower exponent that reflects the different density of the confined field (plasmon) states: $\Delta_{2D} \propto N^{-7/6}$. The shift depends on the inverse size nearly linearly. However, the prefactor dominates at some moderate size of the QD and lessens the LS to 10^{-3} for $N = 100$. The depolarisation is still to be too small to expect experimental consequences.

3. The $\overline{\delta^2}$ considered above the less, the larger the QD size, that is not the case for the deflection due to completely localized modes like in Sec. 1. In this section the localized modes are the surface plasmons of the spherical inclusion (with the dielectric function ϵ_1) in the matrix (with the different dielectric function ϵ_2). $\Delta \propto N^{-2/3}$. Our estimation shows that the level correction, becoming of the order of 50%, plays the important role for the QD of 100 atoms and smaller. We collected all studied contributions to the depolarisation LS and plot them in the log–log scale versus the QD size in Fig. 1.

The depolarisation because of the localized surface QD modes is large enough to propose an experiment supporting our model. It is easy to check that $\overline{\delta^2} \sim \omega_{\text{L}}^{-3}$, which is nearly the frequency of the bulk plasmon in the matrix (with the weak dependence on the mode angular momentum, see [3]). Therefore, changing the optical properties of the matrix surrounding the QD, one shifts the levels. If the bare energy level lies deep in the potential well, its position is nearly independent of the well depth which changes along with the matrix parameters. The deep bare level energy depends only on the well width. Hence, the depolarisation LS is distinguishable from the standard space quantization LS.

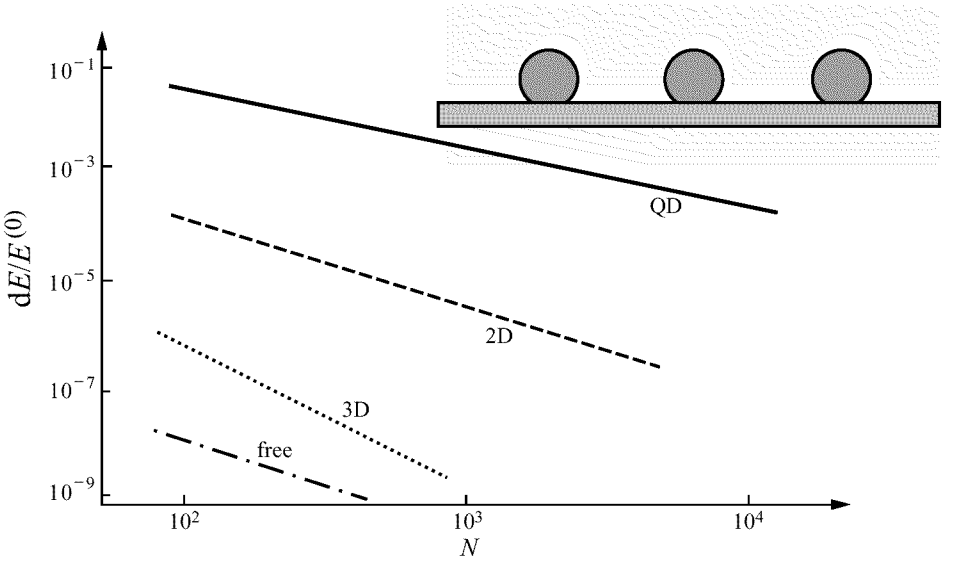

Fig. 1. The level shift of the QD electron, calculated for 4 different depolarisation mechanisms. The slopes and the prefactors of these depolarisation shifts were derived analytically. Inset: The scheme of the QD structure to model.

In summary, the effect of the zero-point oscillations of the free and confined electromagnetic field on the level of the confined electron in the 0D-system, as the closed-shell carbon cluster and the spherical QD, is studied. The depolarisation due to an interaction with the zero-point fluctuations shifts up the bare one-electron state. The gap renormalisation, which follows from the angular momentum dependent LS in the fullerene cluster, is shown to be independent of the fullerene radius. The size dependence of LS in the QDs is different for 4 modes considered in the paper. Although, in general, the depolarisation decreases with the QD size, the **localized** surface electromagnetic mode results in the essential level shift and is to be possibly resolved experimentally for a QD made from some hundreds atoms. Another method to detect the effect could be a measurement of a deep level position of the similar QDs buried by the substrates with distinct optical characteristics.

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